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SUBJECT: Authorization for Release of Technical Information, Control Number: AFRL-PR-ED-**TP**-1998-073
Ingrid J. Wysong (Raytheon) Molecular Models for Reacting Flows: Should Variable Collision Diameters be Used in DSMC Simulations" **EXTENDED ABSTRACT** to be published **(Statement A)**

Molecular Models for Reacting Flows: Should Variable Collision Diameters for Internal States be used in DSMC Simulations? *

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The formulation of expressions for transport properties in nonequilibrium flows has been an active research field for many years [?]. One powerful feature of particle simulation methods such as direct simulation Monte Carlo (DSMC) is that they do not require transport properties as input parameters. Rather, given a sufficiently realistic model of the intermolecular potential and energy transfer, transport properties emerge naturally as a statistical consequence of many collisions along with boundary conditions [?]. Thus, a physically realistic yet computationally tractable model for molecular collisions is of primary importance for DSMC methods.

In a DSMC simulation, collision pairs are selected based on the local density and the velocity-dependent collision cross section. For any given reaction (or relaxation) model, the number of reactions produced in a given calculation (the local reaction rate) will be controlled by the local total collision frequency. Therefore, any increase or decrease in a molecules collision cross section can directly affect its effective reaction rate.

Gorbachev *et al.* [?] have presented a derivation of analytical expressions for the average internuclear distance $R(v,J)$ of a diatomic molecule as a function of vibration and rotation level, based on an accurate internuclear potential. Gimelshein *et al.* [?] have presented an implementation of these expressions in a DSMC code as an addition to the variable soft sphere (VSS) collision model. This intriguing proposal will produce significantly increased collision cross sections for particles with high levels of internal excitation, thus increasing their rates of reaction. Although the expressions for average internuclear distance $R(v,J)$ may be expected to be quite accurate, the effect on intermolecular collision cross section may not be straightforward. The goal of the present discussion is to examine the feasibility of validating the realism of this approach.

Transport properties are an important source of information on intermolecular potentials. The following discussion refers to viscosity, but most of the same arguments apply to diffusion. The viscosity is typically dominated by elastic collisions, so that, in a first approximation, molecules may be viewed as atom-like. If we consider higher-order effects, we must include inelastic collisions. The effect of inelastic collisions on the transport collision integral (for viscosity) is expected to be small (Mason-Monchick approximation), but has not been thoroughly investigated for high temperatures. The other effect specific to molecules is the subject of this paper: that is, higher rovibrational (v,J) states will increase $R(v,J)$, which may in turn increase the collision cross section. Since the population of high (v,J) states will become significant at high temperatures, one might expect that the viscosity at high temperatures may reflect this effect [?]. However, in addition to the difficulty in obtaining accurate viscosity data at very high temperatures, any examination of these data to glean insight into the effect of $R(v,J)$ would need also to disentangle the effect of inelastic collisions.

To estimate the feasibility of validating the effect of increasing $R(v,J)$ at higher temperatures from viscosity data, some estimates are provided for a simple gas of pure molecular hydrogen. The (v,J) populations are in equilibrium and the viscosity collision cross section is given by the VSS cross section where each (v,J) state has a different reference diameter as defined in [4]. Fig. 1 shows that the assumed effect of $R(v,J)$ will begin to significantly decrease the viscosity compared with the VSS model for very high temperatures. However, the effects of dissociation and ionization at these high temperatures in a real gas are expected to be of greater importance to the measurable viscosity than the diameter effect. The calculation indicates that the effect of (v,J) excitation on collision cross section due solely to the diameter as proposed in [4] is likely im-

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possible to verify through viscosity data. However, other effects such as a change in the attractive well depth or inelastic collisions may contribute in reality.

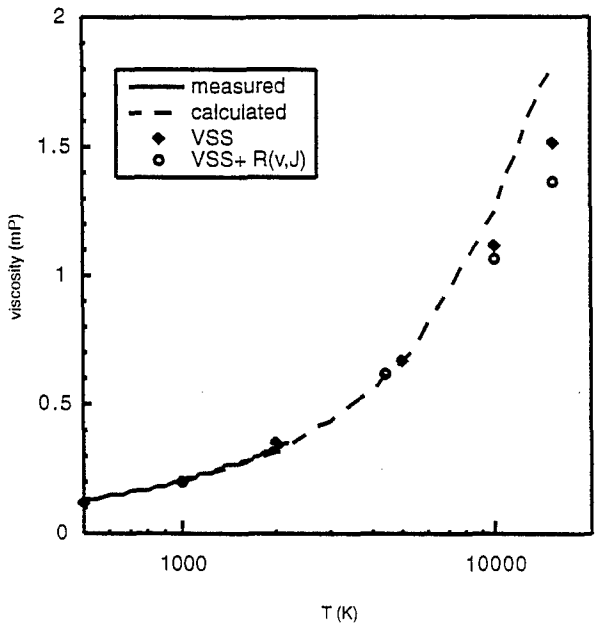

Figure 1: Variation in viscosity due to addition of rovibrational diameters to VSS model

The viscosity effect shown above may seem to indicate that including collision diameters based on *(v,J)* level is of no importance. This conclusion is not necessarily true for highly nonequilibrium gases, which are naturally the regime of interest for DSMC simulations. Certain nonequilibrium gases have highly excited vibrational distributions and, in these cases, an increase in collision diameter for the molecules with high vibrational quantum number could change the chemistry prediction for a DSMC simulation.

A number of experiments using light-induced drift (LID) have been performed which can directly measure the effect of changing *(v,J)* on the diffusion cross section of certain molecules [?]. These measurements show that the diffusion collision frequency, and thus the collision cross section, typically increases a small amount when v is increased from *0* to *1*. On the other hand, these molecules show a decrease in collision frequency by a few percent as J increases. It is not clear how similar the results would be for other molecules. It is also difficult to assess how the results for a small range of *(v,J)* values would change for very high v or J levels. These results do demonstrate, however, that even though *R(v,J)* increases in a predictable way with increasing internal energy, the effect on the intermolecular collision cross section is difficult to predict and is influenced by subtle details of the intermolecular potential.

The proposal to include increased collision cross sections for high *(v,J)* states in DSMC simulations is worthy of examination. It is based on a first-principles approach to fundamental molecular properties. While the majority of flowfields, where the populations of very high internal energy states are insignificant, need not consider such an effect, there are certain nonequilibrium cases where this effect could potentially be important. It seems, however, that the collision cross section may not always increase with *(v,J)* in the straightforward manner proposed. Details of specific characteristics of the applicable intermolecular potential may need to be considered. Some validation of the relationship between internuclear diameter and intermolecular collision cross section is recommended before it is widely applied in DSMC codes.

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